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June 2019 Depleted Uranium Irradiation

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Abstract

Three different irradiations were performed at the Annular Core Research Reactor (ACRR) at Sandia National laboratory on June 4-6, 2019. Each target assembly included two or three depleted uranium targets, as well as a bismuth target and dosimetry packets. In July 2019, the target assemblies were received at TA-48 RC-1, unpackaged, and radiochemical analyses were performed. The analyses included autoradiography and gamma counting of the whole depleted uranium targets, gamma counting of the activation foils, and dissolution of six of the depleted uranium targets, followed by separated radiochemical analysis for a suite of fission products. This report describes the details of the radiochemical analysis and provides final results for the fission and activation product concentrations.

Background

For a series of targets irradiated at the ACRR, it may be possible to collect enthalpy data (measured by thermocouple at the time of the irradiation) and compare this with the energy produced by fission (quantified by analysis of fission product formation in the targets). The goals of this project are to develop an integrated experimental capability for both enthalpy and fission analyses, and ultimately to compare the results of both analyses on a range of material types, including depleted uranium (DU), highly enriched uranium (HEU), and plutonium. Experiments in FY19 have focused on developing the experimental infrastructure and performing an initial comparison of thermocouple and radiochemical analyses of depleted uranium targets irradiated at the ACRR.

Sample Receipt and Unpacking

On July 10, 2019, three different target assemblies were received at TA-48 RC-1. The target assemblies were loaded with depleted uranium targets as well as dosimetry packets, which contained Sc, Ni, Ti, and Fe foils. Each target assembly was housed within a stainless steel canister under vacuum. The targets were disassembled, photographed, and each depleted uranium target sealed in mylar for initial assay by autoradiography (Figures 1 and 2). The dosimetry packets were also removed, and each activation foil individually mounted for analysis by gamma ray spectrometry (Figure 3). C.D. Archuleta and D.T. Olive (MST-16) had originally put together the assembly and were present during disassembly to offer guidance. Tables 1 and 2 provide the sample IDs and masses associated with the different targets and activation foils.
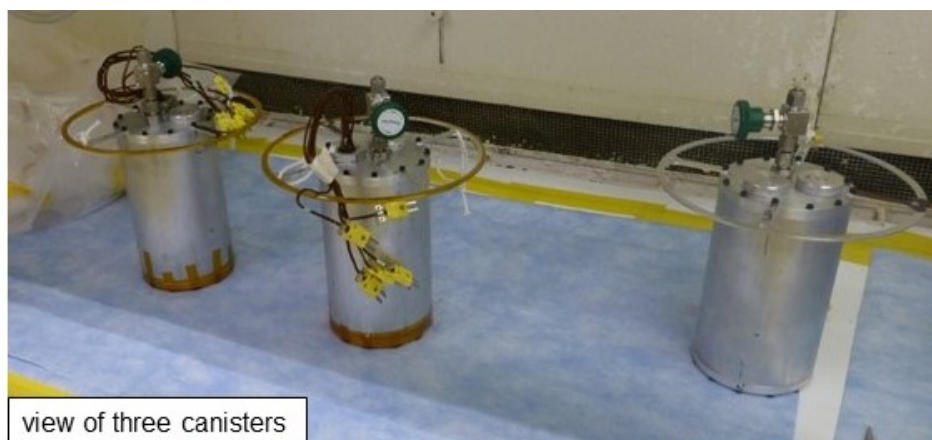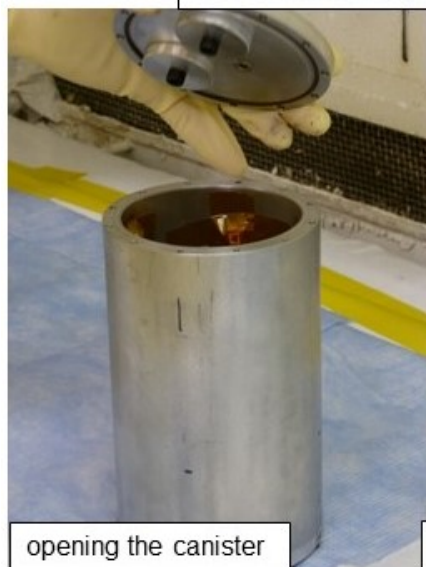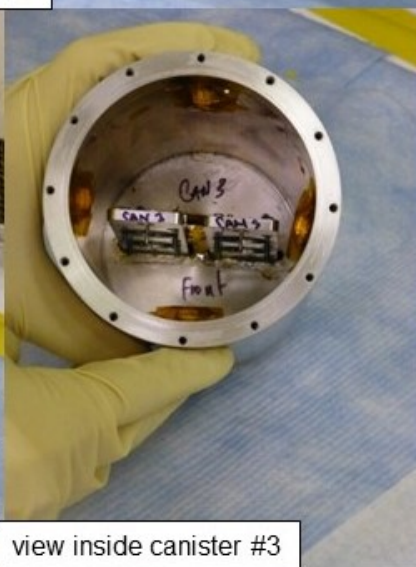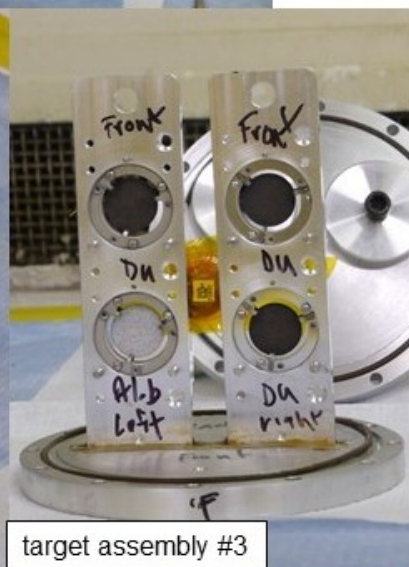
Figure 1. Disassembly of irradiated stainless steel canisters holding the target assemblies.

Figure 2. Sealing depleted uranium targets for initial assay by autoradiography, then mounting for gamma-ray spectrometry.

Figure 3. Mounting activation foils for analysis by gamma-ray spectrometry.

Table 1. Sample ID, target ID and mass of target in grams for seven depleted uranium targets irradiated at the ACRR.

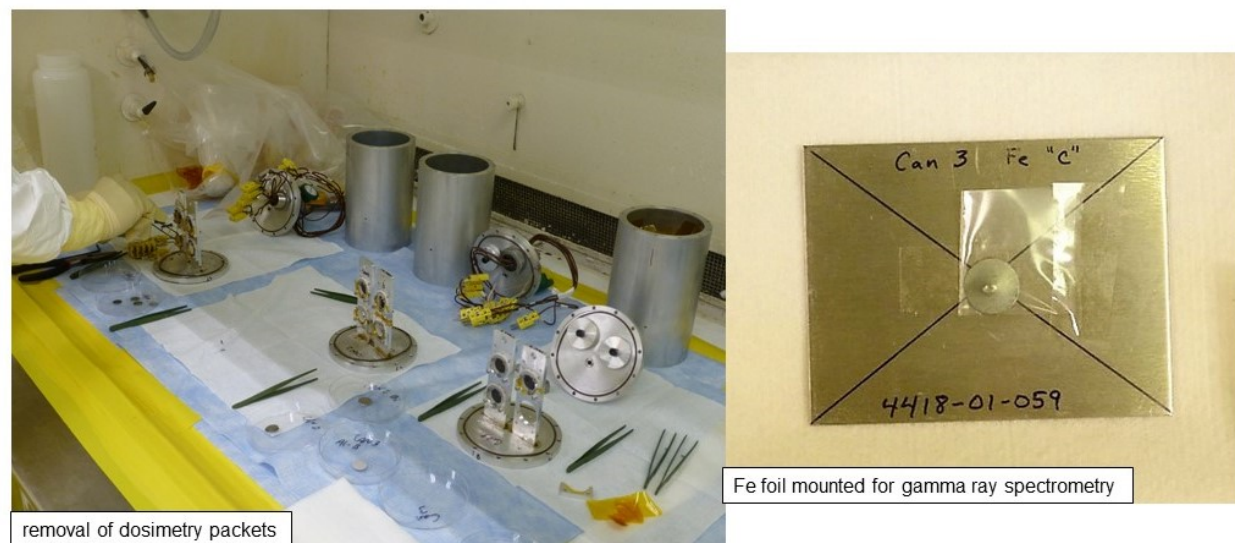| Sample ID	Target ID	Target Mass (g)	Assembly number	Date of Irradiation (2019)	Position in assembly
4416-01	DU-1	3.9827	1	155.8098	Upper left
4416-02	DU-3	4.0796	1	155.8098	Upper right
4417-01	DU-6	4.0399	2	156.7230	Upper left
4417-02	DU-10	3.8032	2	156.7230	Upper right
4418-01	DU-7	3.8965	3	157.6670	Upper left
4418-02	DU-9	3.9097	3	157.6670	Upper right
4418-03	DU-8	4.0219	3	157.6670	Lower right

Table 2. Sample IDs and masses of activation foils associated with the three target assemblies.

Sample ID	Target material	Target Mass (g)	Assembly number	Date of Irradiation (2019)	Position in assembly
4416-01-046	Sc foil	0.0454	1	155.8098	Lower left
4416-01-057	Ni foil	0.2850	1	155.8098	Lower left
4416-01-044	Ti foil	0.1425	1	155.8098	Lower left
4416-01-059	Fe foil	0.1252	1	155.8098	Lower left
4417-01-046	Sc foil	0.0489	2	156.7230	Lower left
4417-01-057	Ni foil	0.2861	2	156.7230	Lower left
4417-01-044	Ti foil	0.1427	2	156.7230	Lower left
4417-01-059	Fe foil	0.1253	2	156.7230	Lower left
4418-01-046	Sc foil	0.0497	3	157.6670	Center
4418-01-057	Ni foil	0.2865	3	157.6670	Center
4418-01-044	Ti foil	0.1426	3	157.6670	Center
4418-01-059	Fe foil	0.1255	3	157.6670	Center

Autoradiography and gamma counting of the whole targets

Autoradiography was performed on all seven irradiated targets using short exposures (10 minutes on the front of each target, 15 minutes on the back). An image of the autoradiography exposures is shown in Figure 4. Although autoradiography is a somewhat qualitative technique, no evidence for non-uniformity or shielding from clips in the target assembly was observed.
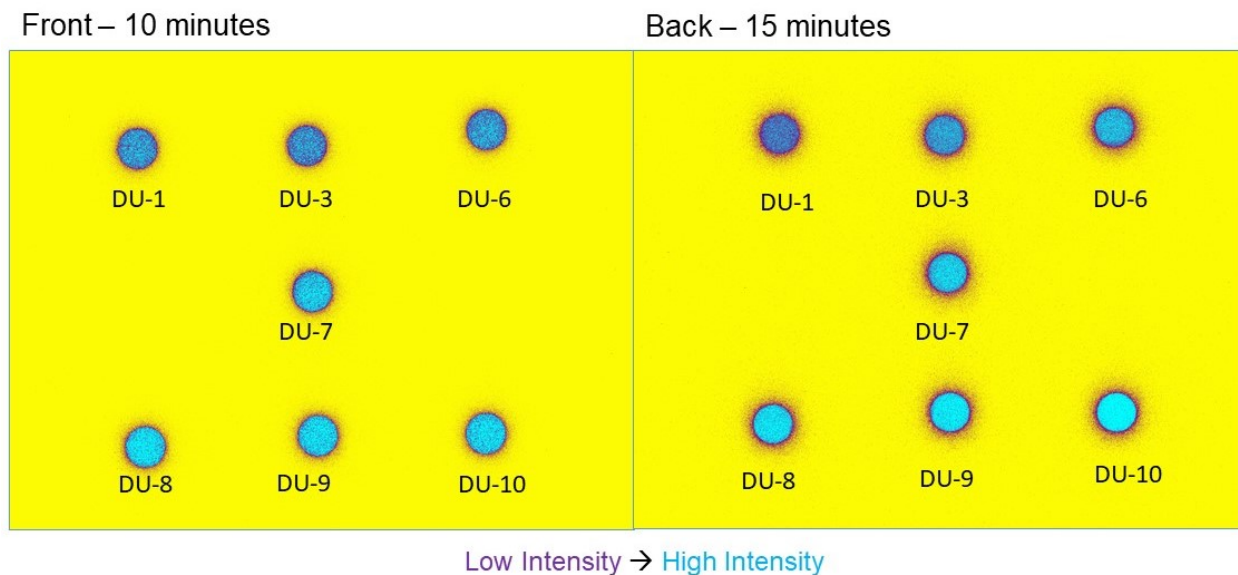
Figure 4. Autoradiography exposures of depleted uranium targets irradiated at the ACRR.

Following autoradiography, each target was mounted for assay by gamma-ray spectrometry. An automated gamma spectrometry counter was used for overnight analysis. Count lengths were 50 to 100 minutes on Counter 76 which is a Li-drifted germanium (GeLi) counter of 14% efficiency. An estimate of the total number of fissions in each target is provided in Table 3 (based on ^{95}Zr).

Table 3. Total fission estimates based on gamma-ray spectrometry of the mounted whole targets.

Sample ID	Target ID	Date of Irradiation (2019)	Initial total fission estimate (fissions/g target)
4416-01	DU-1	155.8098	6.5E+11
4416-02	DU-3	155.8098	6.4E+11
4417-01	DU-6	156.7230	6.4E+11
4417-02	DU-10	156.7230	6.3E+11
4418-01	DU-7	157.6670	6.4E+11
4418-02	DU-9	157.6670	6.1E+11
4418-03	DU-8	157.6670	6.5E+11

Gamma counting of the activation foils

The twelve primary activation foils (Fe, Ni, Sc and Ti) were gamma-counted on 3 counters for 1000 minutes each. Multiple counts (usually 2-4) were done over several days. Multiple isotopes were identified arising from neutron capture and (n,xn) reactions. The average concentrations were estimated based on a weighted average over multiple counts. Those results are shown in Table 4.

Table 4. Results of activation foil counting. The reference date for each measurement is the end of bombardment and uncertainties are reported at $k = 1$.

Foil	Isotope	Can 1 (4416) (atoms/g)	Unc (%)	Can 2 (4417) (atoms/g)	Unc (%)	Can 3 (4418) (atoms/g)	Unc (%)
Fe	Mn-54	9.30E+09	2.4	8.37E+09	2.3	8.30E+09	2.4
	Fe-59	7.45E+09	2.6	6.74E+09	2.4	7.26E+09	2.3
Ni	Co-57	2.56E+08	4.3	2.79E+08	4.9	2.33E+08	6.4
	Co-58	1.36E+11	7.1	1.20E+11	4.3	1.22E+11	2.3
	Co-60	8.74E+08	16.0	8.48E+08	3.5	8.10E+08	2.4
Sc	Sc-46	5.38E+13	2.3	5.18E+13	2.3	5.34E+13	2.3
Ti	Sc-46	1.85E+09	2.7	1.75E+09	2.3	1.60E+09	2.7
	Sc-47	3.33E+09	13.6	2.97E+09	11.1	2.78E+09	17.2

In addition, there were 2 Bi foils and 1 Al-B foil which were included in the cans during the irradiation. Each of these were counted at least twice and the isotopes present were identified and, if possible--quantified. Results are presented in Table 5.

Table 5. Results from Bi and Al-B foils. The reference date for each measurement is the end of bombardment and uncertainties are reported at $k = 1$.

Foil	Isotope	Can 1 (4416) (atoms/foil)	Unc (%)	Can 2 (4417) (atoms/foil)	Unc (%)	Can 3 (4418) (atoms/foil)	Unc (%)
Bi	Ag-110m*	1.26E+08	5.3	1.30E+08	2.9		
	Cr-51*	4.82E+07	12.3	-	-		
	Co-60*	1.48E+08	29.0	-	-		
Al-B	Na-22					1.42E+09	17.3
	Ta-182*					3.49E+10	6.9

Note there were several cases where isotopes (*) were identified which have no obvious precursor in the foil material. Whether these can be attributed to contamination or activation of trace impurities has not been resolved. Additional details and calculations of the activations in the detector foils are located in the appendix.

Dissolution of uranium targets

Four of the targets (DU-1, DU-6, DU-7, DU-8) were selected for initial dissolution on July 11, 2019. Each target was removed from the mylar wrapping and transferred to a Teflon beaker. The targets were immersed in 4 M HNO_3 and dissolved using a slow addition of concentrated HCl. The sample was slowly heated over a period of several hours to drive off excess HCl, and a color change from green to yellow was observed. This color change is consistent with oxidation of any tetravalent uranium species to hexavalent uranium. Ultimately, a final solution of each target (denoted as the "A" solution) was prepared in 4 M HNO_3. These solutions were analyzed directly by gamma-ray spectrometry (whole "A" analysis).

In addition, separated radiochemical analyses were performed on each solution. Dissolution of two additional targets (DU-10 and DU-9) followed a similar procedure and was completed on Friday, July 12, 2019.
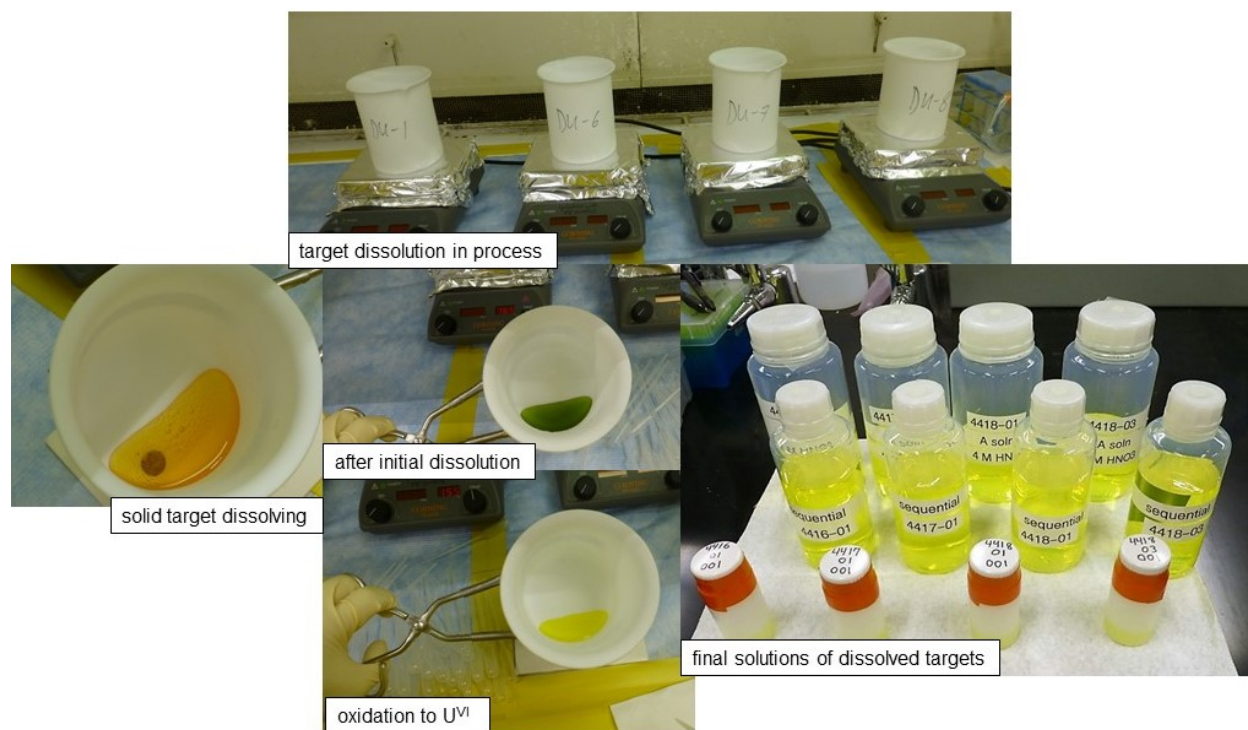
Figure 5. Dissolution of uranium targets.

Separated radiochemical results

A suite of fission product isotopes was measured for the four samples 4416-01 (DU-1), 4417-01 (DU-6), 4418-01 (DU-7) and 4418-03 (DU-8). A single aliquot consisting of ~50% of each sample (accurately weighed) was traced with stable carriers, and a sequential procedure was first used to isolate individual element fractions. These isolated fractions were then further purified for each analyte. Chemical yields were determined gravimetrically, and samples were mounted as chemically pure solid precipitates for analysis by either beta counting or gamma-ray spectrometry. The peak-yield isotopes measured include ^{89}Sr, ^{91}Y, ^{95}Zr, ^{99}Mo, ^{137}Cs, ^{140}Ba, $^{141,\,144}$Ce, and ^{147}Nd. In addition, ^{111}Ag, ^{136}Cs, and ^{156}Eu were measured in the samples, and could provide additional information about the neutron energy during the irradiation. Standard radiochemical procedures were modified to account for both removal of the vast excess of target material and the extent of decay of the shorter half-life fission product analytes (see appendix A).

Tables 6-9 show the results of the fission product analyses. Each radioisotope is reported in units of atoms per gram of target material. The isotopes ^{89}Sr, ^{91}Y, ^{99}Mo, ^{111}Ag, ^{140}Ba, ^{147}Nd, and ^{156}Eu were determined by beta counting. Gamma-ray spectrometry was used to measure ^{136,137}Cs and $^{141,\,144}$Ce. The reference time for each isotope is the irradiation time for the specific target analyzed.

Table 6. Separated fission product results from sample 4416-01 (DU-1). The reference time for all isotopes is the end of bombardment 155.8098 (2019).

Isotope	Atoms/g target	Uncertainty (%)	Measurement method
Sr-89	2.31E+10	3.0	beta
Y-91	3.06E+10	3.5	beta
Zr-95	3.53E+10	5.6	gamma
Mo-99	3.75E+10	2.3	beta
Ag-111	1.86E+08	3.0	beta
Cs-136	2.72E+07	23.8	gamma
Cs-137	3.63E+10	3.2	gamma
Ba-140	3.30E+10	3.2	beta
Ce-141	3.19E+10	5.0	gamma
Ce-144	2.91E+10	5.1	gamma
Nd-147	1.37E+10	4.5	beta
Eu-156	1.81E+08	3.6	beta

Table 7. Separated fission product results from sample 4417-01 (DU-6). The reference time for all isotopes is the end of bombardment 156.7230 (2019).

Isotope	Atoms/g target	Uncertainty (%)	Measurement method
Sr-89	2.38E+10	2.3	beta
Y-91	3.12E+10	2.8	beta
Zr-95	3.79E+10	7.0	gamma
Mo-99	3.77E+10	2.0	beta
Ag-111	1.78E+08	2.1	beta
Cs-136	2.72E+07	11.0	gamma
Cs-137	3.56E+10	2.4	gamma
Ba-140	3.39E+10	2.7	beta
Ce-141	3.22E+10	2.8	gamma
Ce-144	3.08E+10	3.0	gamma
Nd-147	1.37E+10	2.5	beta
Eu-156	1.86E+08	2.4	beta

Table 8. Separated fission product results from sample 4418-01 (DU-7). The reference time for all isotopes is the end of bombardment 157.6670 (2019).

Isotope	Atoms/g target	Uncertainty (%)	Measurement method
Sr-89	2.36E+10	2.4	beta
Y-91	3.11E+10	2.5	beta
Zr-95	3.37E+10	12.5	gamma
Mo-99	3.78E+10	2.0	beta
Ag-111	1.75E+08	2.0	beta
Cs-136	2.83E+07	26.1	gamma
Cs-137	3.51E+10	2.5	gamma
Ba-140	3.26E+10	3.0	beta
Ce-141	3.14E+10	2.8	gamma
Ce-144	2.93E+10	3.0	gamma
Nd-147	1.31E+10	3.5	beta
Eu-156	1.78E+08	2.4	beta

Table 9. Separated fission product results from sample 4418-03 (DU-8). The reference time for all isotopes is the end of bombardment 157.6670 (2019).

Isotope	Atoms/g target	Uncertainty (%)	Measurement method
Sr-89	2.39E+10	2.1	beta
Y-91	3.04E+10	2.5	beta
Mo-99	3.60E+10	2.1	beta
Ag-111	1.80E+08	2.1	beta
Cs-136	2.59E+07	23.6	gamma
Cs-137	3.35E+10	2.2	gamma
Ba-140	3.48E+10	2.1	beta
Ce-141	3.12E+10	2.8	gamma
Ce-144	2.85E+10	2.9	gamma
Nd-147	1.32E+10	2.6	beta
Eu-156	1.78E+08	2.3	beta

Whole gamma "A" solution results

After dissolution, aliquots were dispensed to a standard 20 mL counting vial (Type 2) and counted over a period of about 60 days. Count lengths were 1000 minutes on Counter 76.

The final values determined from a whole "A" solution are taken from a detailed analysis that involves collecting multiple spectra over time, and then performing multiple decay component deconvolution of each gamma line reported. C-NR has developed a spreadsheet-based tool that calculates fissions per unit mass of the "A" solution sample. This spreadsheet includes the necessary physics constants, including half-lives, branching ratios, and fission yields, as well as counter efficiency for that specific peak energy. Decay during bombardment calculations were not performed for this experiment.

The uncertainty is based on the quadrature sum of three components; a fixed efficiency calibration uncertainty, an aliquot weight measure uncertainty, and the intercept uncertainty of however many gamma lines contribute to the final nuclide quantification. The fixed efficiency calibration uncertainty is smaller in this case due to the fact that we calculate a gamma energy efficiency from a mathematical model fitted to a NIST-traceable calibration source. This mathematical model is believed to represent a higher precision estimate of the efficiency as a function of energy.

A critical part of performing the decay component deconvolution is identifying and accounting for interferences. Within the ensemble of gamma lines that are important in the quantification of fission products, there are multiple cases where different nuclides have a gamma line that interferes with (i.e. is within the inherent energy resolution) of a gamma emission line of a different nuclide. In most cases the overlapping contributions are very different in quantity, so that the correction made to the primary nuclide is a few percent or less. In cases where multiple measurements are averaged, the standard deviations of those measurements are included in the overall uncertainty.

The final results of the measurements taken and their associated uncertainties are listed in Tables 10-15 below.

Table 10. Direct "A" solution gamma spectrometry fission product results from sample 4416-01 (DU-1). The mass of the aliquot of "A" solution is 5.747 g. The reference time for all isotopes is the end of bombardment 155.8098 (2019).

Isotope	Atoms/g target	Uncertainty (%)
Zr-95	3.54E+10	2.5
Ru-103	2.28E+10	2.8
Ba-140	4.04E+10	5.3
Ce-141	3.51E+10	2.6
Ce-144	3.55E+10	6.8
Nd-147	1.47E+10	7.6
Cs-137	4.07E+10	4.6

Table 11. Direct "A" solution gamma spectrometry fission product results from sample 4417-01 (DU-6). The mass of the aliquot of "A" solution is 5.764 g. The reference time for all isotopes is the end of bombardment 156.7230 (2019).

Isotope	Atoms/g target	Uncertainty (%)
Zr-95	3.47E+10	3.0
Ru-103	2.19E+10	2.7
Ba-140	3.86E+10	5.5
Ce-141	3.43E+10	2.6
Ce-144	3.34E+10	8.1
Nd-147	1.37E+10	3.2
Cs-137	4.14E+10	9.5

Table 12. Direct "A" solution gamma spectrometry fission product results from sample 4417-02 (DU-10). The mass of the aliquot of "A" solution is 5.733 g. The reference time for all isotopes is the end of bombardment 156.7230 (2019).

Isotope	Atoms/g target	Uncertainty (%)
Zr-95	3.47E+10	2.7
Ru-103	2.18E+10	2.8
Ba-140	4.10E+10	8.1
Ce-141	3.48E+10	2.5
Ce-144	3.21E+10	12.1
Nd-147	1.57E+10	14.6
Cs-137	5.10E+10	10.5

Table 13. Direct "A" solution gamma spectrometry fission product results from sample 4418-01 (DU-7). The mass of the aliquot of "A" solution is 5.716 g. The reference time for all isotopes is the end of bombardment 157.6670 (2019).

Isotope	Atoms/g target	Uncertainty (%)
Zr-95	3.46E+10	2.9
Ru-103	2.18E+10	2.9
Ba-140	3.87E+10	2.4
Ce-141	3.40E+10	2.7
Ce-144	3.42E+10	3.8
Nd-147	1.61E+10	10.5
Cs-137	3.72E+10	10.7

Table 14. Direct "A" solution gamma spectrometry fission product results from sample 4418-02 (DU-9). The mass of the aliquot of "A" solution is 5.760 g. The reference time for all isotopes is the end of bombardment 157.6670 (2019).

Isotope	Atoms/g target	Uncertainty (%)
Zr-95	3.42E+10	2.9
Ru-103	2.18E+10	2.4
Ba-140	4.00E+10	10.4
Ce-141	3.44E+10	2.9
Ce-144	3.51E+10	4.3
Nd-147	1.656E+10	10.9
Cs-137	2.68E+10	23.6

Table 15. Direct "A" solution gamma spectrometry fission product results from sample 4418-03 (DU-8). The mass of the aliquot of "A" solution is 5.737 g. The reference time for all isotopes is the end of bombardment 157.6670 (2019).

Isotope	Atoms/g target	Uncertainty (%)
Zr-95	3.50E+10	3.3
Ru-103	2.21E+10	2.4
Ba-140	4.05E+10	9.2
Ce-141	3.46E+10	2.3
Ce-144	3.05E+10	9.2
Nd-147	1.35E+10	11.1
Cs-137	3.97E+10	9.2

Conclusions and future work

A suite of fission products was successfully measured in six depleted uranium targets irradiated at the ACRR. The fission product data was of high quality, despite a five week delay from the end of the irradiation to receipt at TA-48, RC-1. Comparison of the fission product data with enthalpy measurements is currently underway.

In addition, activation products were measured in a series of detector foils loaded with each irradiation. The activation foils can provide additional information about the neutron spectrum and fluence during the irradiation. Together with the fission products, these isotopes present a detailed picture of the irradiation conditions.

Future work will involve similar irradiations of HEU and Pu metal targets at the ACRR. Efforts are ongoing to identify strategies for removal of milligram quantities of Pu target material from the fission product analytes of interest.

Appendix A.

1. Details of fission product calibrations

Beta counting calibrations

Since the 1950s, radiometric measurements of ^{99}Mo have been used to determine the total number of fissions in a sample. At Los Alamos, this key measurement is performed by beta decay counting chemically purified samples. The ^{99}Mo measurement calibration serves as the basis for fission determinations and has been historically maintained through a series of fission chamber experiments tied to specific gas proportional beta counters.

A K-factor relates the observed beta decay count rate (cpm) of a sample on a specified counter to the total number of fissions in the sample (on a ^{235}U thermal basis, Equation 1, where K_{99Mo} is the K-factor and A_{99Mo} is the sample activity in terms of cpm). First established in the 1950s, LANL K-factors were updated in 1970 in a large experimental campaign coordinated by NIST. Since 1970, the K-factor calibration has been maintained and updated to reflect changes in the particular gas proportional counters and sample mounting. Last updated in the 2000s (to reflect a move to counter 19 and stainless steel backing material), the ^{99}Mo K-factor currently in use is 2.300 x 10^5 ($\pm$1.4%) fissions/cpm (^{235}U thermal basis). This methodology has been recently validated using an external certified ^{99}Mo reference solution.[1]

$$Fissions = K_{^{99}Mo} \times A_{^{99}Mo} \qquad (1)$$

For each radioisotope of interest, a k-factor is maintained that relates the observed beta decay count rate (cpm) of the isotope to the total number of atoms of the isotope in the sample. These k-factors are derived from thermal calibration experiments, the ^{99}Mo K-factor, and the ^{235}U thermal fission yield of the isotope of interest. An example for ^{111}Ag is shown in Equations 2 and 3. Radiochemistry calibrations are regularly performed by the radiochemistry team with irradiated HEU to maintain the validity of this methodology.

$$k_{^{111}Ag}\left(\frac{atoms}{cpm\,^{111}Ag}\right) = \frac{K_{^{99}Mo}\left(\dfrac{fissions}{cpm\,^{99}Mo}\right)}{r_{^{111}Ag}\left(\dfrac{cpm\,^{111}Ag}{cpm\,^{99}Mo}\right)} \times FY_{^{111}Ag}\left(\frac{atoms\,^{111}Ag}{fission}\right) \qquad (2)$$

$$\frac{atoms\,^{111}Ag}{sample} = \frac{observed\ cpm\,^{111}Ag}{sample} \times k_{^{111}Ag}\left(\frac{atoms}{cpm\,^{111}Ag}\right) \qquad (3)$$

For this experiment, the total number atoms of each isotope measured per gram of target material was reported.

Gamma-ray spectrometry calibrations

For gamma-ray spectrometry, all automated counters employed for data collection incorporate a calibrated NIST-traceable source that is counted routinely as part of the sample sequence. The data collected from these sources are routinely evaluated to assure proper operation of each counter.

2. Detailed analysis of the activation foils

Three separate irradiations were performed in the ACRR each with a single can containing multiple samples present. Four activation foils (Fe, Ni, Sc and Ti) were present to enable characterization of the neutron intensity and spectrum. These foils were gamma-counted and eight isotopes were identified among the four foils. These were quantified from the SPECANAL code using the provided irradiation time and among multiple counts (typically 2-4) the average yields and uncertainties at T0 were calculated. In addition, all of the Fe foils also indicate the presence of Cr-51 and Co-60. Both isotopes are present in far higher concentrations than would be expected from (n,alpha) or double neutron capture reactions, respectively. The origin of these isotopes in the Fe has not been resolved.

A simple MCNP model was created using a Watt fission spectrum source transported through 30cm of 0.2 g/cm^3 density of light water. The resulting neutron spectrum provided a starting point to confirm the activation seen in the foils. A second MCNP run was used with each foil set immersed in the neutron source taken from the results of the first run. Based on activation tallies, iterative changes were made to the spectrum to match the foil results. In this methodology, self-absorption of the neutrons was taken into account. The foils were assumed to be 100% pure material and the same spectrum (although with slightly different intensities) was used for all 3 irradiations.

Based on the activation foil results, the estimated neutron fluence for each can was computed and the results are given in Table A-1.

Table A-1. Estimated Neutron Fluence per Can.

	Fluence (n/cm^2)
Can 1	7.5x10^{13}
Can 2	6.8x10^{13}
Can 3	6.8x10^{13}

The computed neutron lethargy spectrum is given in figure A-1.
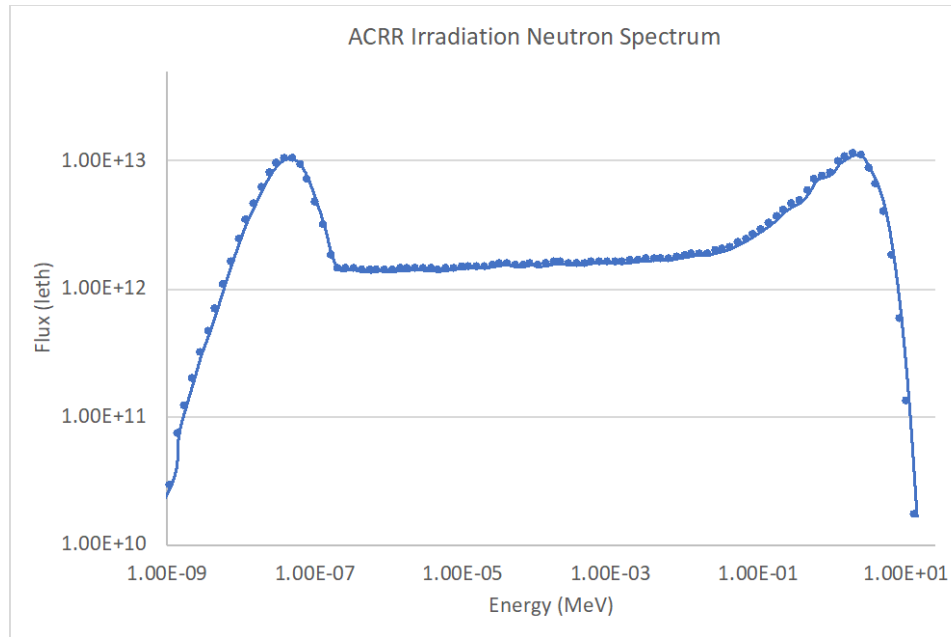
Figure A-1. Neutron lethargy spectrum assumed for all cans.

The detailed results for Can 1, which was given a sample ID of 4416 are given in Table A-2, which give the measured atoms per foil and the MCNP computed results and a C/E (computed over experiment) value for each isotope. Figure A-2 illustrates the atoms/foil and C/E graphically.

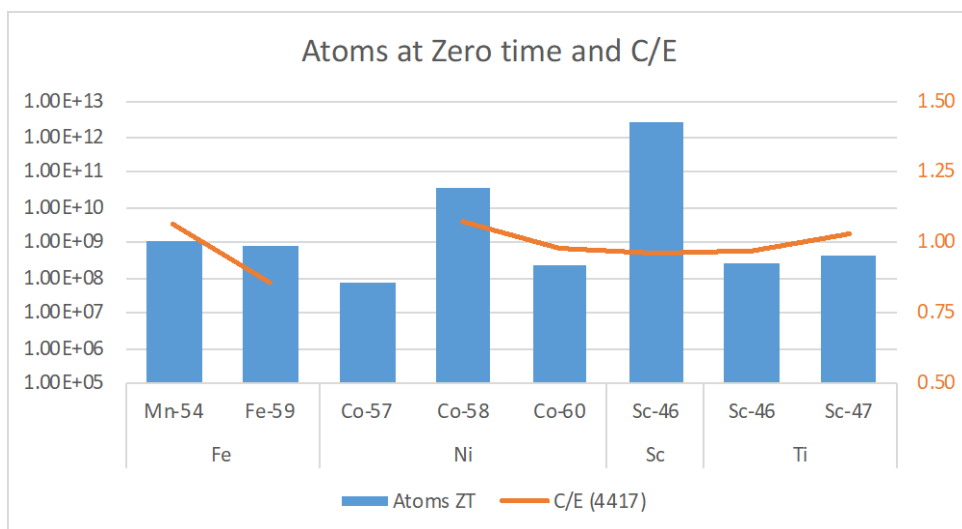**Table A-2.** Can 1 (4416) Detailed Results.

Foil	Isotope	Atoms ZT	Error (%)	MCNP est.	C/E (4416)
Fe	Mn-54	1.16E+09	2.4	1.21E+09	1.04
	Fe-59	9.33E+08	2.6	7.52E+08	0.81
Ni	Co-57	7.29E+07	4.3	3.38E+06	
	Co-58	3.86E+10	7.1	4.02E+10	1.04
	Co-60	2.49E+08	16.0	2.58E+08	1.04
Sc	Sc-46	2.44E+12	2.3	2.48E+12	1.02
Ti	Sc-46	2.64E+08	2.7	2.71E+08	1.03
	Sc-47	4.75E+08	13.6	4.80E+08	1.01
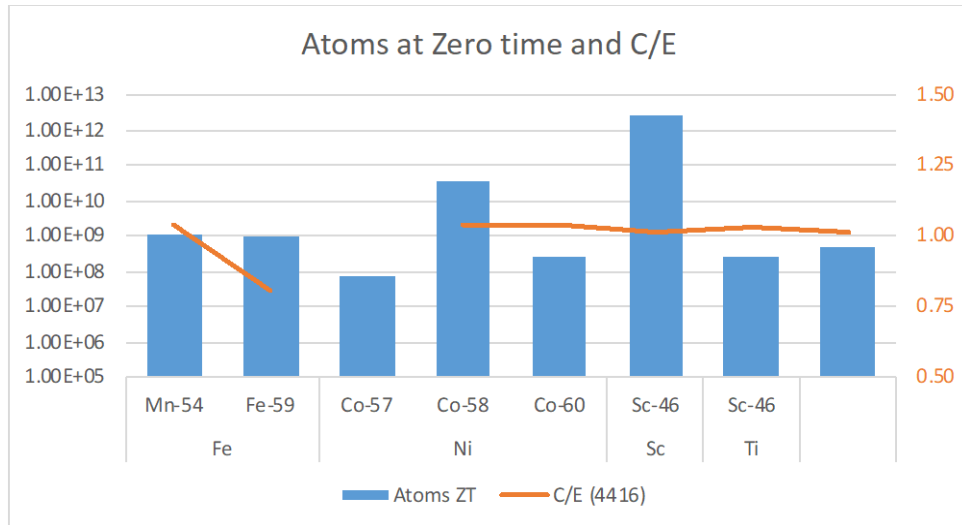
Figure A-2. Results of Can 1 (4416) atoms/foil and MCNP C/E.

**Table A-3.** Can 2 (4417) Detailed Results.

Foil	Isotope	Atoms ZT	Error (%)	MCNP est.	C/E (4417)
Fe	Mn-54	1.05E+09	2.3	1.12E+09	1.06
	Fe-59	8.45E+08	2.4	7.27E+08	0.86
Ni	Co-57	7.98E+07	4.9		
	Co-58	3.43E+10	4.3	3.68E+10	1.07
	Co-60	2.43E+08	3.5	2.38E+08	0.98
Sc	Sc-46	2.53E+12	2.3	2.43E+12	0.96
Ti	Sc-46	2.49E+08	2.3	2.41E+08	0.97
	Sc-47	4.24E+08	11.1	4.36E+08	1.03

Figure A-3. Results of Can 1 (4417) atoms/foil and MCNP C/E.

Table A-4. Can 3 (4418) Detailed Results.

Foil	Isotope	Atoms ZT	Error (%)	MCNP est.	C/E (4418)
Fe	Mn-54	1.04E+09	2.4	1.11E+09	1.07
	Fe-59	9.11E+08	2.3	6.62E+08	0.73
Ni	Co-57	6.68E+07	6.4		
	Co-58	3.50E+10	2.3	3.68E+10	1.05
	Co-60	2.32E+08	2.4	2.38E+08	1.03
Sc	Sc-46	2.65E+12	2.3	2.47E+12	0.93
Ti	Sc-46	2.28E+08	2.7	2.41E+08	1.06
	Sc-47	3.97E+08	17.2	4.35E+08	1.10
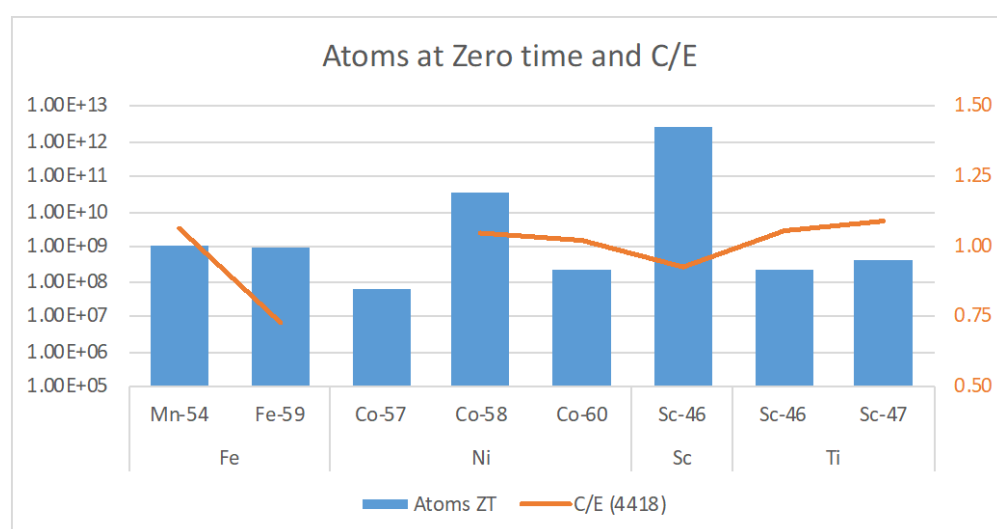
Figure A-4. Results of Can 3 (4418) atoms/foil and MCNP C/E.

The activation results are consistent with MCNP results based on the hypothesized spectra. One outlier is the Fe-59 results which are 20-25% low. This underprediction could be based on more captures in an epithermal resonance. A more rigorous spectrum unfolding would be beneficial. Another outlier is the Ni->Co-57 reaction which was ultimately not included in the analysis. The measured yields (taken from 122 keV peak) are approximately 5-20 times what would be expected from the MCNP calculations.

A comparison of the estimated fissions on the DU foils to MCNP was performed. The U-235 fraction in the DU was not supplied, but a 0.2% atom fraction was assumed based on MCNP calculations. Table A-5 compares the estimates of fissions based on gamma measurements to MCNP results based on spectrum derived from the activation foils.

Table A-5. DU Foil Fission Result Comparisons.

Can	Shot	DU #	Total Fissions	Mass (g)	fiss/g	Average	MCNP	C/E
1	4416	1	2.42E+12	3.9827	6.08E+11	6.08E+11	6.43E+11	1.06
2	4417	6	2.27E+12	4.0399	5.62E+11			
	4417	10	2.25E+12	3.8032	5.92E+11	5.77E+11	5.85E+11	1.01
3	4418	7	2.4E+12	3.8965	6.16E+11			
	4418	9	2.34E+12	3.9097	5.99E+11	5.95E+11	5.85E+11	0.98
	4418	8	2.3E+12	4.0219	5.72E+11			

A good match is seen on the C/E results.

Using these results, we can also use MCNP to compute the relative fissions in U-235 and U-238. The fission breakdown was ~70% U-235, 30% U-238.

3.   Work done in preparation for the irradiation.

During January and February 2019, a series of experiments was performed to optimize and validate methods for fission product analysis from a large DU target mass (chemistry details are provided below). This work demonstrated that up to 2 g of depleted U can be analyzed successfully using an adaption of current methods. Table A-6 displays the predicted feasibility of analysis of fission products of interest from 1 g of depleted uranium (dU) as a function of both the total number of fissions and the number of days after irradiation before counting starts. Counting of separated element fission products typically begins 3-4 days after sample receipt at TA-48. This table was developed using separated element yields obtained from experiments (see details below).

Chemistry methods

In preparation for the June 2019 C7 depleted uranium target irradiation, the radiochemistry team performed an analysis on a 2 g portion of dU metal, a larger mass than our typical experiments. To put this into perspective, a typical irradiation scenario might involve of *ca.* 100 mg of HEU metal producing 10^{14} fissions. Other past experiments at NCERC (National Criticality Experiments Research Center) have involved irradiation of dU and typically generate more than 10^{11} fissions/g and are received within 4 days of sample irradiation. This work was therefore performed to assess any potential problems that might arise from the extra actinide target mass relative to fissions generated than routinely analyzed through standard radiochemical separation methods. The potential problems that were assessed were excess uranium in separated aliquots, low yields of purified fission products and the potential for ^{234}Th interference. ^{234}Th has a 24.10 day half-life and can be an interference in beta counting for isotopes with comparable half-lives.

Fission product analysis from an irradiated target typically involves cold carrier addition for yielding and separated element purifications. Samples are mounted on stainless steel planchets for subsequent counting. Given the low estimated total fissions for the June experiment, an alternative sequential procedure was used. To a solution of 2.27 g of depleted uranium dissolved in 4 M HNO$_3$ were added Ag, Ba, Sr, Cd, Mo, Cs, Zr, Nd, Ce, Y, Tb, Eu, and Sm carriers in acid solution (20 mg each). Each element was then separated sequentially through a combination of wet chemistries and column purifications. The large amount of uranium caused some modifications to be made to existing procedures. ^{234}Th was shown to follow a number of elements through the initial elemental separations but additional separations were developed to successfully remove Th, thus eliminating the potential for

interference. The modified sequential procedure and associated separated element yields were used to develop the predictions shown in Table A-6.

	Days After Irradiation		
	14 days	28 days	14 days
Fission Products	Total Fissions		
	1.64E+11	1.64E+11	1.64E+10
Zr-95	High	High	High
Sr-89	High	High	High
Sr-90	High	High	High
Y-91	High	High	High
Mo-99	High	High	High
Ag-111	High	Medium	Medium
Cd-115	Medium	Low	Low
Cd-115m	Low	Low	Low
Cs-137	High	High	Medium
Ba-140	High	High	High
Ce-141	High	High	High
Ce-143	Medium	Low	Low
Ce-144	High	High	High
Nd-147	High	High	High
Sm-153	Medium	Low	Low
Eu-156	Medium	Low	Low
Tb-161	Low	Low	Low

Analysis Confidence
- High (green)
- Medium (orange)
- Low (red)

Table A-6. Confidence levels for fission products analysis for three different irradiation and time of receipt scenarios, assuming 1 g dU target.

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[1] Berger, J. L. *et. al.* "⁹⁹Mo Calibration Exercises." **2018**, LA-UR-18-29719.